CLAIMS

We claim:

1. A method of preparing a xylene product comprising:

providing a reactor containing a phosphorus-treated ZSM-5-type zeolite catalyst;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of from about 2 hr^{-1} or more and the cofeed of hydrogen is introduced at a hydrogen/hydrocarbon (H_2/HC) molar ratio of less than about 8, and wherein the temperature is from about 500 °C to about 700 °C;

operating the reactor at the start-up conditions for about one-half to about 20 hours; and then

operating the reactor at run conditions wherein the LHSV is from 10 hr^{-1} or less and H_2/HC molar ratio is at least 1.0 and the temperature is from about 500 °C to about 700 °C.

2. The method of claim 1, wherein:

the phosphorus-treated ZSM-5-type zeolite catalyst having a total phosphorus content of from about 0.01 g P/g zeolite to about 0.15 g P/g zeolite.

3. The method of claim 1, wherein:

the start-up LHSV is about 50 or less.

4. The method of claim 1, wherein:

the para-xylene content is at least 90% in the xylene product.

5. The method of claim 1, wherein:

the start-up H_2/HC molar ratio is from about 0.1 to about 8.0.

6. The method of claim 1, wherein:

the reactor is operated at a pressure of from about 10 to about 50 psig.

7. The method of claim 1, wherein:

the toluene/methanol feed has a toluene/methanol molar ratio of from about 1:2 to about 10:1.

8. The method of claim 1, wherein:

the ZSM-5-type zeolite catalyst is treated with at least one of phosphoric acid and ammonium hydrogen phosphate.

9. The method of claim 1, wherein:

the reactor temperature is initially from 200 °C or above and upon introduction of the toluene/methanol feed the reactor temperature is gradually increased at a rate of 1 to 10 °C/min to final start-up temperature from about 500 °C to about 700 °C.

10. The method of claim 1, wherein:

the reactor or catalyst bed inlet temperature is maintained from about 500 °C to about 700 °C.

11. The method of claim 1, wherein:

the catalyst exhibits stable activity for at least 25 hours after start-up of the toluene methylation reaction.

12. The method of claim 1, wherein:

the catalyst has a silica/alumina mole ratio prior to phosphorus treatment from about 25 to about 300.

13. The method of claim 1, wherein:

there is substantially no structural aluminum loss of the catalyst during the toluene methylation reaction.

14. A method of preparing a xylene product comprising:

providing a reactor containing a phosphorus-treated ZSM-5-type zeolite catalyst using a silica/alumina mole ratio of from 25 to 300 prior to phosphorus treatment and a total phosphorus content of from about 0.01 g/g zeolite to about 0.15 g/g zeolite;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of from about 1 to about 50 and the cofeed of hydrogen is introduced at a hydrogen/hydrocarbon (H_2/HC) molar ratio of less than about 8 and wherein the temperature is from about 500 °C to about 700 °C;

operating the reactor at the start-up conditions for about one to about five hours; and then

operating the reactor at run conditions wherein the LHSV is from 50 hr^{-1} or less and H_2/HC molar ratio is at least 5 and the temperature is from about 500 °C to about 700 °C.

15. The method of claim 14, wherein:

the start-up H_2/HC molar ratio is from about 0.1 to about 8.0.

16. The method of claim 14, wherein:

the reactor is operated at a pressure of from about 10 to about 50 psig.

17. The method of claim 14, wherein:

the toluene/methanol feed has a toluene/methanol molar ratio of from about 1:2 to about 10:1.

18. The method of claim 14, wherein:

the ZSM-5-type zeolite catalyst is treated with phosphoric acid or ammonium hydrogen phosphate.

19. The method of claim 14, wherein:

the reactor temperature is initially from 200 °C or above and upon introduction of the toluene/methanol feed the reactor temperature is gradually increased at a rate of 1 to 10 °C/min to final start-up temperature from about 500 °C to about 700 °C, and maintaining the reactor temperature from about 500 °C to about 700 °C.

20. The method of claim 14, wherein:

the catalyst exhibits stable activity for at least 500 hours after start-up of the toluene methylation reaction.

21. The method of claim 14, wherein:

the para-xylene content is at least 90% in xylene product.

22. A method of preparing a xylene product comprising:

providing a reactor containing a phosphorus-treated ZSM-5-type zeolite catalyst using a silica/alumina mole ratio of from 25 to 300 prior to phosphorus treatment and a total phosphorus content of from about 0.02 g/g zeolite to about 0.13 g/g zeolite;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of from about 1 to about 50 and the cofeed of hydrogen is introduced at a hydrogen/hydrocarbon (H_2/HC) molar ratio of less than about 8, and wherein the temperature is from about 500 °C to about 700 °C;

operating the reactor at the start-up conditions for about one to about two hours; and then

operating the reactor at run conditions wherein the LHSV is from 5 hr^{-1} or less and H_2/HC molar ratio is at least 2 and the temperature is from about 500 °C to about 700 °C; and wherein

the catalyst exhibits stable activity for at least 500 hours after start-up of the toluene methylation reaction.